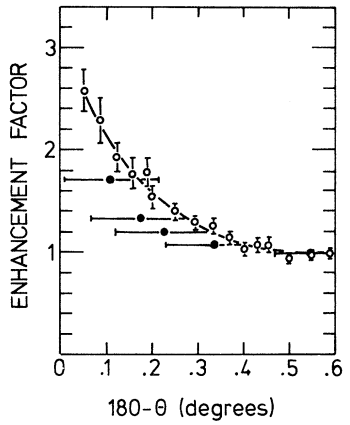

FIG. 3. Enhancement factor $F(\theta)$ as a function of observation angle for 1-MeV He backscattering from Pt at a depth $z = 75$ Å. Open circles, theory; full circles, data (Ref. 1) taken with ~ 100–200 Å depth resolution and representing maximum enhancement factors.

observations of enhanced yields in backscattering at 180° without need to consider neither particular distributions of atoms in the medium (those involving long- or short-range order), nor the electronic disturbances produced by the projectile in its incoming path.

One of us (R.A.B.) would like to acknowledge discussions with O. Crawford and O. Auciello on the subject of this work. This work was supported in part by the Argentine Secretaría de Estado de Ciencia y Tecnología.

Note added.—After this manuscript was completed the authors were made aware of similar work by O. H. Crawford; see Phys. Rev. Lett. 44, 185 (1980).

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Observation of Matthiessen's Rule in Strong Magnetic Fields

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(Received 17 April 1979)

Measurements of electrical resistivity as a function of temperature reveal that deviations from Matthiessen's rule (DMR) in aluminum can, under suitable conditions, be made to vanish in a strong magnetic field. Theories explaining DMR on the basis of anisotropic and inelastic scattering predict such behavior suggesting that such scattering is the source of DMR in dilute aluminum alloys at low temperatures.

In this Letter we present evidence for the existence in aluminum of a magnetic field and temperature range in which the electrical resistivity obeys Matthiessen's rule. We believe that is the only known domain in which this holds[1] and that its existence sheds light on the source of deviations from Matthiessen's rule (DMR) in metals.

The number of theories proposed over the years to explain DMR is huge. Bass,[2] in his review article, lists fourteen different *categories* of theories. At present, none of these theories is completely satisfactory for all temperatues and impurity concentrations. If we ignore those theories which invoke changes in the Fermi surface or other such gross effects (they cannot explain the DMR observed in very dilute alloys) the list of proposed low-temperature theories includes those which consider many-body effects,[3] low-temperature phonon drag,[3] interference between phonon and impurity scattering,[4] inelastic impurity scattering,[5] and small changes in the phonon spectrum (see Ref. 2 for a complete list of references). However, the most successful of the recent theories are those which take into account the anisotropy and energy dependence of the electron-phonon interaction.[6] These theories are the

only ones which have been reasonably successful at predicting DMR over a wide temperature range and most theoretical work in this field now takes this approach. There has been, however, considerable doubt whether such effects are actually the principal cause of DMR, particularly in the light of the nonsaturation of the DMR in the extreme dirty limit.[7] The measurement of the electrical magnetoresistivity in a high magnetic field is a method for clarifying this issue.

The theories of DMR based on the nature of the scattering mechanisms make specific predictions concerning the behavior of the magnetoresistivity. Foremost is the prediction that under special conditions the total magnetoresistivity will be the sum of the separate magnetoresistivities for each scattering mechanism. This is equivalent to a vanishing of the DMR. The conditions are the following[8]: First, the electric and magnetic fields must be crossed. Second, the electron orbits must be closed and must each possess at least twofold symmetry about the magnetic field direction. Third, the magnetic field must be sufficiently strong that electrons on each orbit make at least several revolutions before being scattered (high-field limit). Last, the metal should be uncompensated.

The vanishing of DMR arises from the effect of the magnetic field on the electron distribution function. In the absence of a magnetic field, isotropic elastic impurity scattering and anisotropic inelastic phonon scattering compete to produce a compromise distortion of the electron distribution function. This causes the total resistivity to be greater than the sum of the separate resistivities expected for each scattering mechanism; the result is DMR. As a magnetic field is applied, the field and the scattering mechanisms compete in their effect on the electron distribution function, and at high fields under conditions of sufficient symmetry the nature of the scattering mechanisms ceases to play any role at all. In this case the distortion of the distribution function is determined solely by the geometry of the electron orbits. Because of this, the total conductivity is strictly the sum of the individual conductivities for each scattering mechanism. In uncompensated metals, such as aluminum, the strict additivity of individual magnetoresistivities follows directly[9] and amounts to a vanishing of DMR.

To test this prediction we measured the resistance and magnetoresistance of aluminum-silver alloys. Aluminum was chosen because (1) it has a high Debye temperature, which allowed us to take data over a wide temperature range while still remaining in the high-field limit; (2) it is cubic, which allowed us to choose from among a number of symmetry axes; (3) its Fermi surface is generally believed to be closed; and (4) it is readily available in high purities, is easily alloyed, and single crystals are simple to grow. Its disadvantages are a possible open orbit[10] (for $\vec{H} \| [100]$), the often-observed magnetic breakdown and linear magnetoresistivity, and the fact that the third-zone electron surface does not have the requisite symmetry. This latter point is not crucial since it may be shown that at high fields the third zone contributes less than 2% to the total resistivity.[11]

Silver was chosen as the impurity for a number of reasons: It has a low natural occurrence and high solubility in aluminum; it has an atomic volume near that of aluminum; it causes an increase in ρ_0 per atomic percent that is sizable[12]; and dilute aluminum-silver alloys exhibit large DMR's in zero field.[2,7]

The alloy crystals were manufactured from zone-refined aluminum having a residual resistance ratio (RRR) of 15 000. A 0.2% master alloy was made using zone-leveling methods. The specimens were then produced by successive dilutions of the master alloy with pure material, followed by zone leveling and crystal growing. From the large single-crystal, specimens were cut with an acid saw and a chemical crystal-facing machine. Chemical slicing and facing techniques were employed rather than spark abrasive cutting since we wished to fabricate specimens with minimum internal damage. There is considerable evidence that the linear term in the electrical magnetoresistivity of aluminum is substantially enhanced by such damage; since this term is not well understood we wished to minimize it in our specimens.[13] The silver content of the alloys spanned a range up to 165 ppm, corresponding to an RRR of 150.

Orientation of the crystals was accomplished via the usual back-reflection Laue x-ray techniques. The crystals were oriented with $\vec{H}$ parallel to the [110] direction to within a few tenths of a degree. This direction was chosen to avoid the possible open orbit in the [100] direction,[10] and because a survey of the literature revealed that the linear magnetoresistance appeared to be minimal when $\vec{H}$ is parallel to [110] and no oscillatory magnetoresistance is present.[14] (A disadvantage of minor consequence is that a small number of the second-zone orbits do not have the

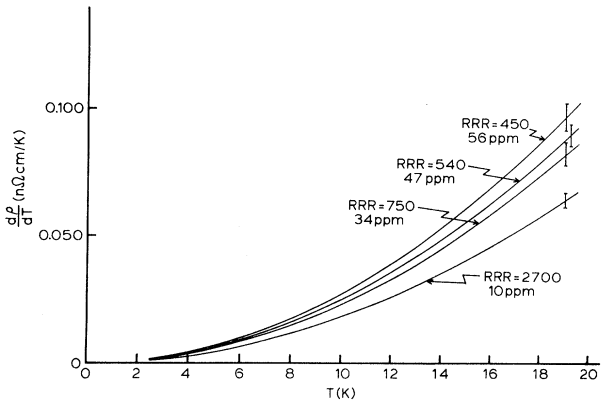

FIG. 1. Temperature derivative of the resistivity of aluminum alloys vs temperature in zero magnetic field.

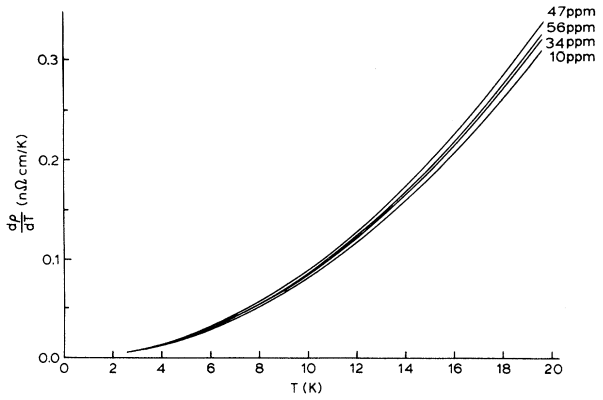

FIG. 2. Temperature derivative of the resistivity of aluminum alloys vs temperature in an 8.3 T field.

requisite symmetry.)

The specimens were mounted in a jig designed to minimize thermal stress as the system was cooled. The electric current was injected through brass electrodes soldered to the ends of the crystal with a "high" resistivity solder. This high-impedance current injection and extraction is absolutely necessary to avoid probe effects.[15] The voltage was measured with a Keithley nanovoltmeter; the specimen temperature was monitored with a calibrated germanium thermometer and a magnetic-field-insensitive capacitance thermometer, the latter being used as a transfer standard.

Before turning to the data, we observe that there are two ways to write Matthiessen's rule[1]: (1) As stated by Matthiessen and Vogt,[16]

$$\frac{d\rho_{\text{alloy}}(c_1,T)}{dT}=\frac{d\rho_{\text{pure}}(0,T)}{dT}, \tag{1}$$

or, (2) as is usually done, $\rho_{\text{alloy}}(c_i,T)=\rho_{\text{pure}}(T)+\rho_0(c_i)$, where $\rho_0(c_i)$ is the residual resistivity. For our purposes, Eq. (1) is more useful. In general, it is not possible to obtain a sufficiently pure specimen to determine accurately $\rho_{\text{pure}}(T)$, and, for aluminum in particular, $\rho_{\text{pure}}(T)$ and $\rho_{\text{alloy}}(T)$ can have very different temperature dependences because of various breakdown effects.[17] By use of Eq. (1) we essentially avoid having to measure a pure specimen; that is, we assume Matthiessen's rule is obeyed if

$$\frac{d\rho_{\text{alloy}}(c_1,T)}{dT}=\frac{d\rho_{\text{alloy}}(c_2,T)}{dT}. \tag{2}$$

Accordingly, we present our results in this manner in Figs. 1 and 2.

Figure 1 shows the temperature derivative of the resistivity in zero magnetic field for four alloys ranging from 10 to 56 ppm. Figure 2 displays similar results for an 8.3 T field. Data for the more dilute alloys (less than 6 ppm) are not shown due to the prominence of magnetic breakdown, which alters the temperature dependence of the resistivity.[17] For the less dilute alloys (greater than 70 ppm), the high-field regime could not be unambiguously attained at the highest fields available. Consequently, data for these alloys are also not shown. We have used the usual least-squares-fitting procedures to obtain these curves and a conservative estimate of the error in the derivatives is that it is less than 5%. We have shown such an error in Fig. 1 (for $T \simeq 19$ K) for reference. In Fig. 1 we see the expected large DMR; the curves rapidly diverge as the temperature increases. This indicates that the temperature-dependent portion of the alloy resistivity is strongly dependent on purity. The observed deviations scale approximately as the residual resistivity ratio as expected.[7] However, as may be observed in Fig. 2, in a strong magnetic field most of the deviations have disappeared, to within the error inherent in the experiment. We attribute the remaining small differences in the curves to measurement and alignment errors. We notice that the curves in Fig. 2 do not vary with concentration in any systematic manner; in fact, the 47 ppm curve is now highest.

We have observed, therefore, a regime in which Matthiessen's rule is obeyed. We have argued that the theories that invoke anisotropic and inelastic electron-phonon scattering to ac-

count for DMR in zero magnetic field also explain the absence of DMR in a strong magnetic field, under the conditions established in these experiments. This provides strong evidence that the anisotropy and energy dependence of electron-phonon scattering is the principal cause of DMR in aluminum, at least at low impurity concentrations.

This work has been supported by the Research Corporation and by the University of Cincinnati Research Council. One of us (D.K.W.) is grateful to C. Herring for first bringing to his attention the fact that conductivities due to different scattering mechanisms can be strictly additive in a strong magnetic field, a fact that underlies this work. We are pleased to acknowledge the advice of B. Addis in sample preparation and helpful discussions with F. P. Esposito, B. W. Maxfield, J. W. Wilkins, and J. C. Garland. W. R. Datars supplied us with some very pure aluminum used in some of the experiments.

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Microscopic Basis of Miedema's Empirical Theory of Transition-Metal Compound Formation

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(Received 26 November 1979)

The success of Miedema's empirical theory of transition-metal compound formation has given credence to the physical picture used in its construction. Self-consistently calculated electron densities, state densities, and heats of formation reveal this picture to be inappropriate. Miedema's success is shown to result from the implicit incorporation of a dominant chemical trend, which is well described by Pettifor's d-bond model.

The success of an empirical theory due to Miedema and co-workers[1] has generated interest in the microscopic mechanisms responsible for intermetallic compound formation. The theory asserts that a large body of experimental data reflects the interplay of *just two* constituent properties, a rather ill-defined "electronegativity", $\varphi(z)$, and the electron density, $\rho(z)$, at the boundary separating atomic cells in the constituent (z is the atomic number). The heat of compound

